junction biased on the first zero-field step to resonant motion of a fluxon; and the linewidth of the radiation to thermal fluctuations in the fluxon velocity. Second-order effects such as plasmon-fluxon interaction, fluctuation in the fluxon profile, and geometrical irregularities have not been included because of the smallness of the fluctuations. We have found excellent agreement between the theory and experiments even on short junctions, suggesting that the theoretical result Eq. (16) with the absence of model parameters is more general than is its derivation and applicable also on short junctions. The discrepancy (less than a factor of 2) is probably due to external noise entering via bias current or temperature fluctuations.

The samples were fabricated at the University of Salerno, Italy, and we acknowledge the fruitful cooperation with the Josephson group there. This work was supported in part by the Danish Natural Science Research Council, the Danish Ministry of Education, Consiglio Nazionale delle Ricerche, Italy, and by the Ansaldo Company, Genova, Italy.

[a]Permanent address: Institute of Radio Engineering and Electronics of the Academy of Sciences of the U.S.S.R., Marx Avenue 18, Moscow GSP-3, U.S.S.R.

[b]Permanent address: Istituto di Fisica, Università di Salerno, I-84100 Salerno, Italy.

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Role of Spin-Nonconserving Forces in the Critical Dynamics of Fe at the Curie Point

F. Mezei

Institut Laue-Langevin, F-38042 Grenoble Cédex, France, and Central Research Institute of Physics, 1525 Budapest, Hungary

(Received 3 May 1982)

Modern, high-resolution neutron-scattering techniques, in particular neutron spin echo, were applied to reinvestigate the critical dynamics of iron at temperatures above the Curie point. It was found that previously claimed evidence for a crossover was in error, and that the dynamical scaling hypothesis unexpectedly breaks down at small wave numbers. This breakdown is shown to be due to spin-nonconserving forces, which are tentatively identified with the thermodynamic random fields.

PACS numbers: 75.40.Fa, 05.70.Jk, 75.50.Bb

The nature of the transition between the ferromagnetic and the paramagnetic states at the Curie point of the most common ferromagnet, iron, is still an object of controversy. The problem has attracted attention since decades ago and subsequent investigations raised as many questions as they answered.

In magnetism research neutron scattering plays a pivotal role by offering the only way of direct, model-independent measurement of the static and dynamic spin-spin correlation functions. However, in many cases insufficient resolution makes the neutron scattering data limited to a too narrow band, as was the case with the previous investigations of the dynamics of critical fluctuations in iron. Other microscopic experiments can often probe a wider range, but they present the inherent risk of all nondirect measurements: They can easily be misinterpreted by use of a wrong model. The extraordinary progress of neutron scattering techniques in the last decade has extended the resolution range by several orders of magnitude. This is why the present reinvestigation of the critical dynamics of iron at temperatures above the Curie point $T_C \simeq 770\,^\circ\mathrm{C}$ has been undertaken with use of two of the novel methods, neutron spin echo (NSE)[1] and advanced time-of-flight (TOF) spectroscopy. It is little surprise that the results prove wrong both certain extrapolations of previous, lower-resolution neutron scattering data, and suggestions based on nondirect evidence.

In the dynamical study of critical fluctuations we are concerned with the lifetime of these fluctuations. This information is contained by the Fourier-transformed spin-spin correlation function $S(q,\omega)$, which is directly related to both the neutron scattering cross section and the generalized susceptibility function,[2] and from all evidence it can be given in the $(\vec{q},\omega)$ domain of our interest in the form

$$S(\vec{q},\omega)=S(q)\Gamma_q/(\Gamma_q{}^2+\omega^2). \tag{1}$$

The object of the study is to determine the energy width Γ_q as a function of the wave number q.

One of the fundamental predictions of dynamical scaling theory[3] is that at $T=T_C$

$$\Gamma_q=Aq^z, \quad z=(5-\eta)/2, \tag{2}$$

where A is a constant, and the Fisher exponent η is expected to be a small number between 0 and 0.1. Previous neutron scattering data[4,5] gave $z=2.7\pm0.3$ in the range $0.05<q<0.3$ Å^{-1}. However, anomalies recently observed in hyperfine-field relaxation experiments have been interpreted[6] as evidence that, with decreasing q, z crosses over to a value close to 2. The present results show that in fact these anomalies have another origin and Γ_q follows the power law (2) with $z=2.48\pm0.05$ down to $q\simeq0.01$ Å^{-1} without any sign of a crossover.

Another fundamental theoretical prediction[3,7] is that at temperatures $T>T_C$ and for $q\to0$ (in the so-called hydrodynamic limit)

$$\Gamma_q=Dq^2, \tag{3}$$

where the spin-diffusion constant D vanishes as a function of the temperature as $T\to T_C$:

$$D=D_0\tau^y, \quad \tau=(T-T_c)/T_c. \tag{4}$$

Previous neutron scattering experiments[4,8,9] have been evaluated with the assumption (3) and contradictory values were obtained for y. The present experiment shows that Eq. (3) is valid neither in the $q\to0$ limit, nor in any substantial range of q, and that the dynamical scaling behavior does not apply for small q values. This is a very unexpected result, since Eq. (3) is a simple, general consequence[7] of spin conservation characterizing the exchange Hamiltonian. It will be shown that the observed strong breakdown of spin conservation is not due to the theoretically much investigated dipole-dipole interaction.[10-12] I conclude instead that it corresponds to the $3d$-spin-lattice relaxation due to the thermodynamic random forces provided by the spin-orbit interaction. The dipolar, anisotropy, etc., effects would be expected to become dominant as T approaches T_C and thus to lead to crossover in the critical exponents. In contrast, random forces can be shown to produce merely a perturbation-like anomaly, which vanishes at $T=T_C$ and masks the scaling behavior at small q's for $T>T_C$, exactly as observed. In sum, just to the contrary of what was believed, at $T=T_C$ there is no evidence for any deviation from the dynamics expected for the exchange model, but a marked perturbation sets in above T_C. In what follows I will present the experimental results and the arguments which lead to these conclusions.

The sample used was 99.9995% pure, 2-mm-thick iron plate, polycrystalline with large crystallites of 1–3 mm dimensions. T_c has been determined with a relative precision of 0.05 K in the NSE experiment and 0.1 K in the TOF experiment. The resolution of the TOF spectrometer used, the IN5 instrument at the Institut Laue-Langevin, was 18 μeV full width at half maximum. In the paramagnetic-type[1] NSE work no full use could be made of the 0.01-μeV resolution capability of the IN11 instrument,[13] since the q range was limited by beam geometry to $q>0.006$ Å^{-1}.

In the data-reduction procedure $S(q)$ in Eq. (1) was assumed to have the well-known Ornstein-Zernike form, $S(q)\propto\chi_T/(1+q^2/\kappa_1{}^2)$, where κ_1 is the effective inverse coherence length, and χ_T the thermodynamic susceptibility. Thus, the Fisher exponent η was effectively taken as 0, but it was checked that its choice between 0 and 0.2 has no effect on the Γ_q values obtained. κ_1 displays a critical temperature dependence[14]: $\kappa_1\simeq1.22\tau^{0.69}$ Å^{-1}, which was found to be in good agreement with the present scattering spectra. Standard self-consistent TOF data-reduction procedures were applied, and it was established that the corrections these implied were small in all cases. The error of final linewidths Γ_q has been estimated to be about 10%.

Figure 1 shows the measured Γ_q values at $T \simeq T_C$, together with previous results. The straight line represents $Aq^{5/2}$ with $A = 130$ meV $\text{Å}^{5/2}$ which is seen to give a very good fit over 4 orders of magnitude in Γ_q. For technical reasons the NSE data were taken at $T_C + 0.2$ K, and not at T_C, which explains why these points lie somewhat below the line. Furthermore, the 0.1-K uncertainty of T_C in the TOF experiment is not quite a negligible source of error. Taking this into account one finds that the most probable value of z is 2.48 ± 0.05, instead of $z = 2.5$ shown in the figure.

The results at temperatures $T > T_C$ are shown in Fig. 2. The various discontinuous lines were calculated from a fully empirical interpolation formula, which will be introduced below [Eq. (6)]. It is conspicuous that at no temperature do the data correspond to the expected Dq^2 law (straight line of 45° slope) over any sizable range of q. Instead, as $q \to 0$, Γ_q tends toward a constant value Γ_0, which varies with the temperature and vanishes at T_C. This is the most unexpected feature found in the present work. Even the best previous results[7] were limited by modest resolution to wave numbers $q \geq 0.03$ Å^{-1}, and could therefore be force-fitted by the Dq^2 law. Nevertheless, for $q \geq 0.03$ Å^{-1} the agreement is excellent between the present and previous results.

The dynamical scaling hypothesis,[3] i.e., the existence of a scaling function $f(x)$ such that

$$\Gamma_q(T)/\Gamma_q(T_C) = f(\kappa_1/q), \tag{5}$$

is examined in Fig. 3. The various symbols show the experimental values of $\Gamma_q(T)/\Gamma_q(T_C)$ derived from the results in Figs. 1 and 2, while the continuous line represents the function $f(x)$ calculated for Ising ferromagnets.[15] It is seen that the scaling hypothesis (5) does not hold on the whole, but data points corresponding to relatively high wave numbers ($q > 0.03$–0.05 Å^{-1}) do indeed fall on a common line close to the predicted one. This is why previous studies appeared to confirm the theory.[15] The present results can be well described by the purely empirical formula

$$\Gamma_q(T) = Aq^{5/2} f(\kappa_1/q) + B\kappa_1 g(q). \tag{6}$$

Here the first term is the scaling contribution of Eq. (5), where $A = 130$ meV $\text{Å}^{5/2}$ and the conveniently chosen scaling function $f(x) = 0.4284x^{1/2} + \exp(-0.4284x^{1/2} - 2.4x + 0.7x^2 - 0.2x^4)$ is similar to that of Ref. 15, but with a somewhat deeper minimum at $x \sim 1$. In the second term, describing the nonscaling, anomalous contribution, $B \simeq 0.075$

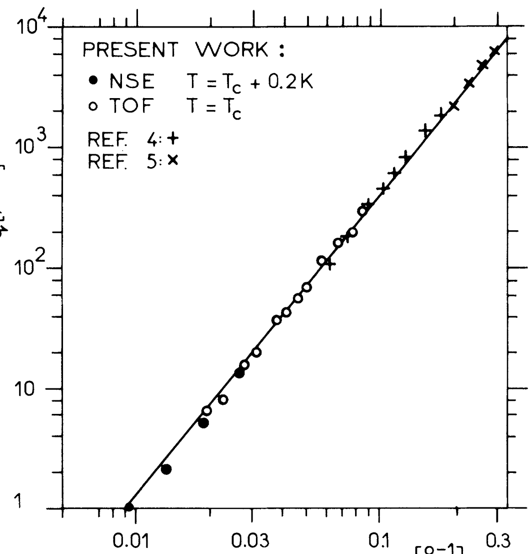

FIG. 1. Present and previous results on the wave-number dependence of the spin-relaxation rate at $T = T_C$.

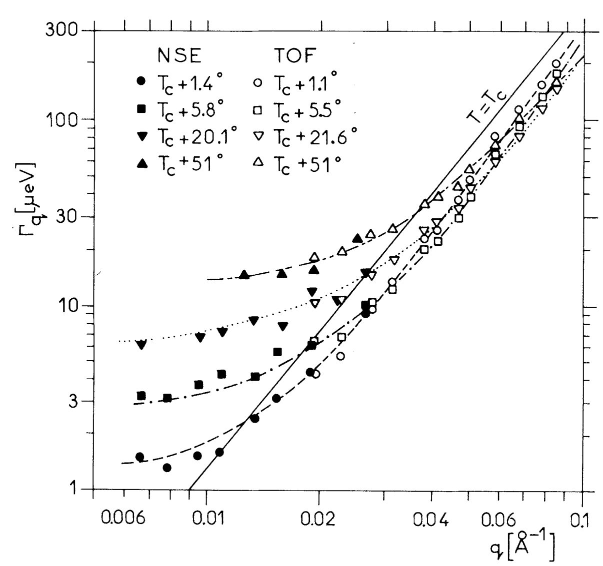

FIG. 2. Measured relaxation rates at various temperatures. For comparison the straight line is the same as in Fig. 1. The discontinuous lines were calculated from Eq. (6) for $T - T_C = 1.5$, 5.6, 20, and 51 K, respectively.

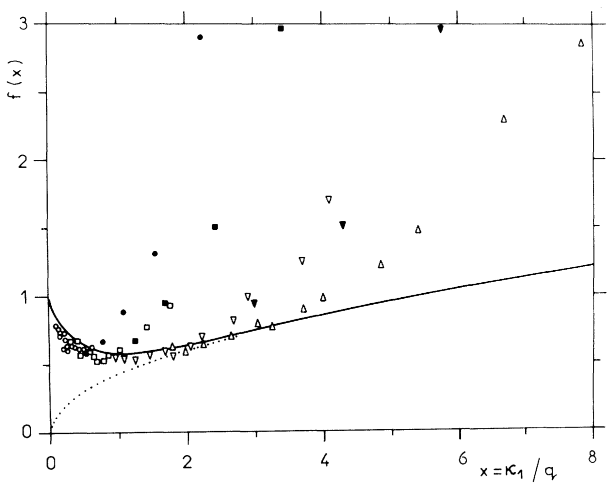

FIG. 3. Comparison of the measured $\Gamma_q(T)/\Gamma_q(T_C)$ values with the Resibois-Piette scaling function (Ref. 15, continuous line). The dotted line is proportional to $\sqrt{x}$, and the symbols are those in Fig. 2.

meV Å and $g(q)$ is a cutoff function which can be chosen, e.g., as $g(q)=1/(1+q^4/q_c^4)$ with $q_c=0.03$ Å^{-1} in order to get a good fit to the experimental results, as shown in Fig. 2.

The above-mentioned hyperfine-field anomalies[6] correctly indicated the breakdown of Eq. (5), and it turns out that they are fully accounted for by Eq. (6). They were misinterpreted[6] by assuming arbitrarily that for low q values and close to T_C another scaling function would apply, i.e., a crossover from one behavior to another would occur. Figure 3 clearly shows that the breakdown of scaling is not of this type: The anomalous points do not show any tendency to converge to a common line.

The theoretical predictions on dipolar effects in Refs. 10 and 11 are neither qualitatively nor quantitatively compatible with the present findings. It was suggested[12] that in these studies demagnetization was omitted, which, together with the anomalous term in Eq. (6), would reduce considerably the dipolar contributions. Thus, no manifestation of the dipolar interaction is expected at $q>0.0005$ Å^{-1} and/or $T>T_C+1$ K.

Crystal anisotropy was predicted[16] to give rise to a $\Gamma_0 \propto \kappa_1^{-1/2}$, which is, however, incompatible with the present findings. Relaxation due to the Van Vleck pseudodipolar interaction has not yet been theoretically investigated, and cannot be ruled out as a possible explanation.[17] This short-range interaction should not affect the data at $T=T_C$, but it can become important at high temperatures. Unfortunately, the approximate theories used to evaluate the real dipolar effects are inadequate for this much stronger interaction.[18]

On the other hand, it is easy to show by using standard theories (such as in Ref. 10) that an interaction of the form $\sum h_i S_i$ could explain the observed approximate proportionality between Γ_0 and κ_1. By symmetry the h_i's must average to zero, and they could just correspond to the relatively slowly fluctuating part of the thermodynamical random fields, responsible for the equilibrium between the spin system and the lattice. Of course, a rigorous theoretical analysis remains to be carried out in order to see if this Brownian-motion–type model is feasible, and if there are other possible explanations.

The author is indebted to Ph. Nozières and H. Capellmann for very stimulating discussions, and to P. Dagleish and M. Serve for valuable technical assistance.

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